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Intercomparison of Results of Several Receptor Models for Apportioning Houston Aerosol

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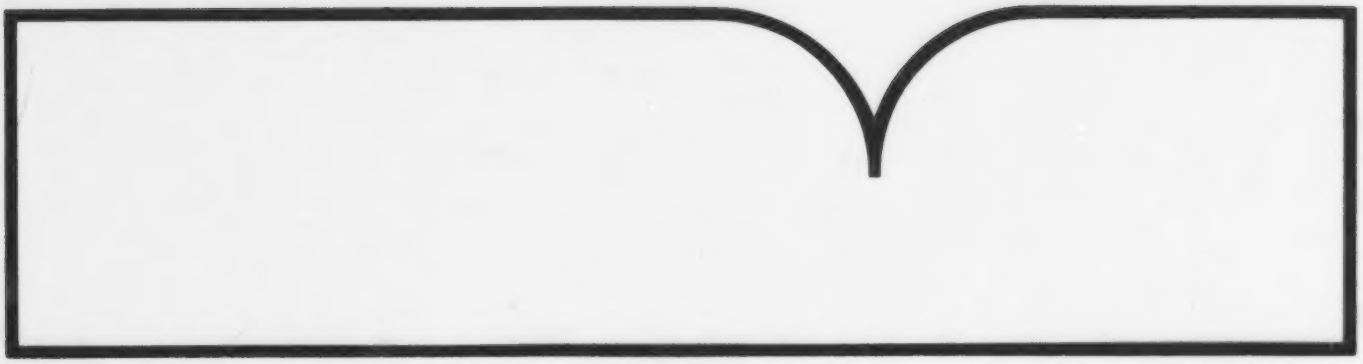

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16. ABSTRACT

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INTERCOMPARISON OF RESULTS OF SEVERAL RECEPTOR MODELS FOR APPORTIONING HOUSTON AEROSOL

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Using a set of data on aerosol composition in Houston, TX, nine participants of the Quail Roost II receptor modeling workshop estimated impacts of major classes of emission sources. The data set included mass, elemental composition, and ionic composition of fine- and coarse- fraction aerosol collected at the University of Houston during 18 12-hr periods from Sept 10 to 19, 1980. Methods used to analyze this data set included chemical mass balance, ridge regression, multiple linear regression, and target transformation factor analysis. Some participants performed additional analyses using x-ray diffraction, scanning electron microscopy, and light microscopy. The number of classes detected ranged from 4 (by multiple linear regression) to 23 (by light microscopy). The results were condensed into eight general categories, intercompared, and found to agree favorably. Differences in results were associated with the use of different source profiles.

INTERCOMPARISON OF RESULTS OF SEVERAL RECEPTOR MODELS FOR APPORTIONING HOUSTON AEROSOL

Introduction

At the recent Quail Roost II Workshop, participants used synthetic and real data sets to evaluate the comparability of several types of receptor models.[1] Here we report the intercomparison based upon the real data set, which consisted of analytical results for ambient aerosol samples collected at the University of Houston in Houston, TX.[2] The sampling site was near grassy areas and campus buildings to the west and south, a large paved parking lot 60 m to the north, and a major freeway 0.9 km to the northeast. Refineries were located to the east 5 km and beyond. Industrial districts were located 5 km to the northeast, east, south, and southwest.

Four dichotomous samplers were operated simultaneously from Sept 10 to 19, 1980, to collect ambient aerosol in the fine (0 to 2.5 μm) and coarse (2.5 to 15 μm aerodynamic equivalent diameter) size ranges. The filters were changed twice daily (at 0600 and 1800 CST). Table I summarizes the analyses of each type of filter. Five weeks before the workshop, participants were given data on mass, carbon, and ionic and elemental composition for 18 consecutive sampling periods. Table II shows the average concentration of each species. For particles smaller than 4 μm, the ratio $^{14}C/^{12}C$ was measured at the National Bureau of Standards and used to deduce the fraction of carbon from fossil fuel combustion.[3]

Filter samples were sent to some participants who performed further analyses. Crystalline species were determined by x-ray diffraction (XRD) analysis at Lawrence Berkeley Laboratory (LBL),[4] and the resulting XRD spectra were also analyzed by Davis at the South Dakota School of Mines (SDSM).[5] Individual particles from each coarse-fraction sample were analyzed by scanning electron microscopy with automated image analysis and x-ray spectrometry (SAX) at the State University of New York (SUNY) at Syracuse.[6] Individual particles from one coarse-fraction sample were analyzed by light microscopy (LM) by Crutcher of Northwest Microlab.[7] The compositions of particles from emission sources in Houston were not available. Thus, it was necessary to use source signatures measured elsewhere; participants were provided with libraries compiled by Watson[8] and Cass and McRae.[9] Also provided were the compositions of three Houston soil and road dust samples, and two Houston emission inventories.

Receptor Model Methods and Results

Except as noted below, participants analyzed the coarse and fine fractions separately. Six participants based their receptor model calculations on elemental and ionic compositional data; one participant used carbon and XRD data; one used SAX data; and one used LM data. No specifications were given for the categories to be used in reporting results. Consequently, the various participants used about 40 different ones. To facilitate comparisons, the participants' results were condensed into eight general categories: marine, crustal, vehicle exhaust, steel, nonferrous metals, sulfate, carbon, and nitrate. The latter three were defined to consist of sulfate, carbon, and nitrate in excess of their amounts in the other five categories. The sulfate category included the related cation, ammonium. Table III presents the participants' results in the eight categories for aerosol collected during daytime, Sept 10.

Source Apportionment Using Ionic and Elemental Composition

Effective Variance Chemical Mass Balance [CMB(EV)]. Shah of Environmental Research and Technology (ERT-A) used the CMB(EV) method developed by Watson.[1,8] A library of about 25 source profiles from Watson[8] and Cass and McRae[9] was used. Included were a profile for noncatalyst car emissions consisting of 21% lead and another for a weighted mixture of car and truck emissions containing 13% lead. For some samples, the weighted mixture was selected to represent the vehicle exhaust category; for others, the noncatalyst car profile was used. Solutions were obtained using six to nine profiles in each of the fine- and coarse-fraction samples. For each sample, fine-fraction sulfate was designated to be either ammonium sulfate or ammonium bisulfate (but not both).

To test for effects of subjectivity in the choices of marker elements and source profiles, Heisler of Environmental Research and Technology (ERT-B), independently followed a procedure similar to that of Shah. For any particular sample, ERT-A and ERT-B did not always use the same set of marker elements and source profiles. For example, ERT-B always used both ammonium sulfate and ammonium bisulfate as profiles for sulfate in the fine fraction, whereas ERT-A always used only one of them. The consequence of this difference was very small. However, the meaning of the vehicle exhaust category is ambiguous because a source profile describing a single vehicle type (noncatalyst cars) was used interchangeably with a profile describing a mixed population of cars and trucks.

De Cesar, Cooper, and Core of NEA, Inc. used a CMB(EV) method similar to that of ERT-A and ERT-B. A library of 33 source profiles, including the three Houston road dust and soil profiles, was used. Most of the other profiles were taken from the Portland Aerosol Characterization Study. Sulfate and ammonium were fit with single-species profiles. The profile selected to represent the vehicle exhaust category in the fine fraction pertained to a weighted mixture of cars and trucks; its lead content was 14%. The number of profiles used in the least-squares fits ranged from six to eight for the fine fraction and from six to nine for the coarse fraction.

Weighted Ridge Regression Chemical Mass Balance [CMB(RR)]. Williamson, Balfour, and Schmidt of Radian Corp. applied both weighted least-squares and weighted ridge regression methods to the Houston data set in a manner described in References 1 and 10. Computations were made using the average daytime and nighttime concentrations measured between 0700 Sept 11 and 0700 Sept 19. The source profiles used in the analysis were taken from Watson[8] and Cass and McRae.[9] The weighting coefficients consisted of uncertainties in the ambient concentrations but not in the source profiles. For both the least-squares and CMB(RR) methods, 18 profiles were used.

Radian's CMB(RR) results for 18 components were less affected by multicollinearity than their least-squares results. The concentrations predicted by CMB(RR) were positive numbers for almost every source, whereas about 35% of the concentrations predicted by the least-squares method were negative. Moreover, the predicted standard errors σ were smaller with CMB(RR) by factors of typically 2 to 5 or more. Nonetheless, the case of 18 profiles was such a severe test of CMB(RR) in terms of multicollinearity that the reported concentrations exceeded 2σ for only two sources in each size fraction. This suggests that optimal performance would have been obtained if fewer than 18 profiles had been used.

Multiple Linear Regression (MLR). Dattner of the Texas Air Control Board (TACB) analyzed Houston aerosol using a backward stepwise unweighted multiple linear regression (MLR) method.[1] Factor analysis indicated four significant multielement components in each size fraction. In each size fraction, two components were determined to be significant by MLR. In the fine fraction, the tracer species for those components were sulfur and bromine; in the coarse fraction, they were silicon and calcium. It was assumed that sulfur and bromine were representative of the sulfate and vehicle exhaust categories, respectively, and that silicon and calcium together represented the general crustal category. The small number of samples in the data set prevented the detection of additional components that significantly affected the variance in mass. At least 40 samples are needed for MLR analysis, but the Houston data set contained only 18.

Target Transformation Factor Analysis (TTFA). Hopke, Severin, and Chang of the University of Illinois (UI) applied TTFA to Houston aerosol.[1,11] Statistical tests suggested the presence of four detectable components in the fine fraction. Their mean concentrations during daytime on Sept 10 and the names given them are: 17.6 $\mu g/m^3$ vehicle exhaust, 35.2 $\mu g/m^3$ sulfate, 0.04 $\mu g/m^3$ smog, and 1.0 $\mu g/m^3$ refinery. Because the sulfate, smog, and refinery components consisted of mixtures of sulfate, nitrate, and carbon, in Table III we present these components as a sum of all three mixtures. In the coarse fraction, five components were detected: their mean concentrations on Sept 10 are shown in Table III with the soil and limestone components combined in the general crustal category. These results do not reveal the full capabilities of TTFA. As in the case of MLR, at least 40 samples are needed, but only 18 samples were provided to workshop participants.

Source Apportionment Using XRD, SAX, and LM Data

Effective Variance Chemical Mass Balance Using XRD Data [CMB(XRD)]. Davis of SDSM used a CMB(EV) method similar to that used by investigators from ERT and NEA.[1,8] However, the only elemental data used were the carbon concentrations. The other data used were XRD results for the following crystalline species: ammonium sulfate, lead ammonium sulfate, quartz, calcite, dolomite, and halite. These XRD results consisted of Davis' analysis of XRD spectra provided by LBL. Davis determined the relative abundances of the crystalline species by applying a self-normalization procedure[5] that requires that the results accounted for the total measured mass minus carbon. Profiles consisting of the abundances of seven species were constructed for the following five sources: vehicle exhaust, combustion, ocean aerosol, soil, and road dust. The combustion profile contained the equivalent of 12.3% lead (expressed as 12.7% lead oxide). The soil and road dust profiles were obtained by XRD analyses at SDSM of the road and soil samples from Houston. The fine and coarse ambient concentrations were combined into a single fraction for which the CMB computations were performed. Results for the combustion category are listed in Table III as the sum of the sulfate and carbon categories. Subsequent to the CMB calculations, the contributions of transportation and combustion to the fine and coarse fractions were made on the basis of their major components (e.g., sulfate in combustion) present in both size fractions.[12] The results reported here do not reveal the full capabilities of CMB(XRD) because the samples were too lightly loaded for complete XRD analysis to be performed.

Q-Mode Analysis of SAX Data. All coarse-fraction ambient samples, as well as source materials used in the apportionment, were characterized by SAX under the same operating conditions to produce "signatures." These signatures consisted of the weight percent contribution by each of 25 particle types. A fixed classification scheme was used for all particles regardless of particle size. The source signatures used in the apportionment covered a variety of area and point source types. Most were obtained in and around Syracuse, NY, but the three Houston soil and road dust samples were included. Using the supplied Houston emissions inventories, 30 source signatures were chosen to represent the Houston area.

All 18 ambient and all 30 source sample signatures were combined in a matrix and subjected to a Q-mode factor analysis[13] with a VARIMAX rotation. The extremes in variable space (25 particle classes) were identified. No Houston ambient samples represented extreme points in variable space after the VARIMAX rotation. This indicated that all important source types were in the data set. "Extreme" samples were those closest to the rotated factor axes and at the outer boundary of the hyperellipsoid. The source apportionment consisted of a vector solution for ambient samples contained within the boundaries of the hyperellipsoid defined by the 16 most extreme signatures. The composition score on the 16 axes was reported as the fractional contribution of each source type to each ambient sample. Products of the fractional contributions and the masses measured by β-gauge were then used to represent the results in units of $\mu g/m^3$.

Light Microscopy (LM). One coarse-fraction ambient sample on a Nuclepore filter was analyzed using LM.[1,7] Particles were first identified in 23 different classes. Then, on the basis of densities assigned to the classes and measured particle dimensions, the sample's composition was constructed. The mass concentration of each particle type was determined by multiplying the sample's total mass concentration by the particle type's fractional mass abundance. Table IV shows the results for ten specific crustal categories; in Table III, these specific categories are combined into the general crustal category. The remaining 13 detected categories were condensed into general categories in the following way: the steel category was used for industrial hematite, rust, iron scale, and magnetite spheres; the sulfate and carbon categories were used for soot, tire wear, charred wood, plant parts, pollen, and secondary industrial particles.

Discussion

Total Mass

When the Houston data set was first obtained, the measured mass was apportioned to the measured species plus the oxygen and hydrogen assumed to relate to them.[2] The amounts that could be accounted for in the fine and coarse fraction were 78 ± 10% and 98 ± 20%, respectively. Because measured species plus assumed oxygen and hydrogen do not account for all fine-fraction mass, the way that mass is used in the calculations will affect the prediction of mass. Three of the investigators (ERT-A, ERT-B, and NEA) did not use mass as an explicit parameter in their CMB(EV) computations. Their results for the fine fraction agree well with each other but account for only 70 to 80% of the measured fine-fraction mass, which is consistent with the fraction accounted for by the measured species.

Both MLR and TTFA used mass explicitly as a parameter fit by least-squares procedures. Consequently, the TTFA results for average mass are in almost exact agreement with the average measured values for both the fine and coarse fractions, and the TTFA results agree fairly closely with the measured value for the particular sampling period shown in Table III. MLR permitted an intercept; its values were -0.1 and -2.5 $\mu g/m^3$ for the fine and coarse fractions, respectively. The intercepts plus predicted component values agree closely with the measured values. CMB(XRD) used mass implicitly in normalizing the abundances of seven species so that their sums in the fine and coarse fractions equaled the total mass. Thus, the CMB(XRD) results agree fairly closely with the total mass.

Vehicle Exhaust Category

Because lead is the most important marker for the vehicle exhaust category, the concentration predicted for this category will be related closely to the abundance of lead in the source profile. The abundances of lead in source profiles used by different participants varied by a factor of 4, causing estimates for the vehicle exhaust category (Table III) to vary by a similar factor. Such variations partly reflect the use of different definitions of the category. For example, for the results shown in Table III ERT-B used a profile for noncatalyst cars and ERT-A used a profile for a mixed population of vehicles, which caused their results to differ by 44%.

In the CMB(EV) analysis performed by NEA and ERT-A, the abundance of lead in the vehicle exhaust profile was 13-14%; this profile was assumed to represent a mixed population of vehicles. Such values differ greatly from the 4 to 5% deduced by MLR and TTFA. The former is based on source measurements made before 1979 and may overestimate the lead content in 1980. Also, the former includes primary emissions only. Because the MLR and TTFA profiles are based on ambient measurements, they may include the actual lead content plus secondary aerosol associated with vehicles. If the vehicle exhaust profile deduced by TTFA could be proven to be correct, then substantial increases in the CMB(EV) estimates for the contributions of vehicles to ambient air would be necessary.

According to Table III, the CMB(XRD) result for the vehicle exhaust category is much lower than the results by other methods. However, such a large difference pertains only to a single 12-hour sampling period on September 10, 1980 and may be anomalous. When all sampling periods between September 10 and 18 were considered, the average result for vehicle exhaust obtained by CMB(XRD) agreed well with the results by other methods.[1]

Crustal Component

Table IV shows how the crustal component, which constitutes at least 70% of the coarse-fraction mass concentration, was resolved into specific components by XRD, SAX, and LM. For quartz and calcite, the XRD data by LBL are in fairly good agreement with the LM results. Because the sample loadings were light, XRD could not detect the clay component reported by LM. The numbers of crustal components determined by the various methods are: ten by LM, eight by SAX, five by XRD, and two by CMB(EV), MLR, TTFA, and CMB(XRD). It is encouraging to note that SAX and LM together resolved about 12 $\mu g/m^3$ or 40% of the crustal component into the following specific

source classes: cement, slag, fly ash, industrial calcium oxide and carbonate, lime kiln, and glass. Good agreement existed between LM and SAX for the first three of these specific classes but not for the last three. Only SAX could resolve fly ash into components from coal and oil.

Conclusions

For the eight general categories, the source apportionment results obtained by the different methods show fairly good agreement. Although apportioning aerosols into eight categories is of great value, obtaining categories that are more specific to emission source types would be desirable. The sulfate, nitrate, carbon, and crustal categories were defined by their chemical nature rather than by emission source type. Thus, it is encouraging to note that LM and SAX did successfully resolve the general categories into several specific source related categories. By combining the CMB results with the LM and SAX results, it is possible to resolve the coarse fraction mass into 15 specific categories.

Although the present study provides valuable insight into the relative performances of several receptor models, certain limitations in the study prevented a demonstration of the models' full capabilities. One limitation was the lack of source profiles specific to Houston. Most were obtained in other cities, making their applicability to Houston only an assumption. Three soil and road dust profiles for Houston were provided, but it is not known if these were representative of the mineral dust suspended in Houston's atmosphere. A second limitation was the lack of a protocol for reporting the vehicle exhaust category. This led some participants to include only primary emissions from noncatalyst cars and others to include primary emissions from a weighted mixture of vehicles. A third limitation was that the 18 sampling periods were too few for MLR and TTFA, which require at least 40 samples. A fourth limitation was that the sample loadings were too low for the full capability of XRD to be demonstrated.

Overall, this intercomparison was valuable because it was the first of its kind and provided impetus to better define the data requirements of receptor models. Recently, a program of ambient and source sampling designed to overcome the limitations of the Houston data set was completed in Philadelphia. Ambient samples were collected during 60 12-hr sampling periods between Jul 15 and Aug 13, 1982. Also, several local source signatures and numerous soil samples were obtained. The data will be available by Sept 1983 and may be used in a comprehensive test of receptor models.

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TABLE I. SUMMARY OF SAMPLING DEVICES AND ANALYTICAL PROCEDURES USED TO DETERMINE AEROSOL COMPOSITION IN HOUSTON FROM SEPTEMBER 10 TO 19, 1980

Sampler Type	Manufacturer (Model)	Height of Inlet, m	Filter Medium	Species Measured	Analytical Procedures
Dichotomous	Sierra (244)	2	Teflon[a]	NO_3^-, SO_4^{2-} H^+ NH_4^+	Ion chromatography Gran titration Colorimetry
Dichotomous	Beckman	4	Teflon[a]	Mass Elemental composition Crystalline species	β-gauge X-ray fluorescence X-ray diffraction
Dichotomous	Beckman	4	Nuclepore[b]	Elemental composition Individual particles	X-ray fluorescence Light microscopy (LM) SAX[d]
Dichotomous	Beckman	4	Quartz[c]	C	Combustion
Hi vol (<4 μm)	Prototype	2	Quartz[c]	C ^{14}C	Combustion β-counter

[a] 1 μm pore size (Membrana Corp.).

[b] 0.3 μm pore size (Nuclepore Corp.).

[c] QAST 2500 (Pallflex Corp.).

[d] Scanning electron microscopy with automated x-ray analysis.

TABLE II. AVERAGE COMPOSITION (ng/m^3) OF HOUSTON AEROSOL IN FINE AND COARSE FRACTIONS FOR 0700 SEPT 10 TO 0700 SEPT 19, 1980[a]

Species	Fine	Coarse
Mass	38600 ± 300	24800 ± 200
Al	123 ± 60	1093 ± 366
Si	210 ± 32	2990 ± 965
S	4834 ± 279	397 ± 430
Cl	32 ± 6	366 ± 111
K	119 ± 6	170 ± 20
Ca	155 ± 8	2775 ± 144
Ti	-8 ± 9	36 ± 12
V	-2 ± 5	-5 ± 6
Cr	4 ± 3	7 ± 3
Mn	14 ± 2	21 ± 3
Fe	162 ± 9	604 ± 34
Co	1 ± 2	0 ± 2
Ni	4 ± 1	4 ± 1
Cu	28 ± 3	18 ± 3
Zn	84 ± 5	58 ± 4
As	2 ± 3	0 ± 2
Se	1 ± 1	0 ± 0
Br	55 ± 3	36 ± 3
Rb	0 ± 1	1 ± 1
Sr	1 ± 1	8 ± 1
Cd	0 ± 2	0 ± 2
Sn	2 ± 3	-1 ± 3
Sb	6 ± 3	-2 ± 3
Ba	48 ± 13	91 ± 16
Pb	465 ± 23	124 ± 10
SO_4^{2-}	14610 ± 1207	907 ± 165
NO_3^-	593 ± 617	1629 ± 235
NH_4^+	4180 ± 379	<190
Na	317 ± 254	278 ± 238
C	7100 ± 467	3100 ± 36

[a] The tabulated results are based upon final β-gauge analysis in Sept 1980, ionic analyses in Sept and Oct 1980, carbon analyses in Oct and Nov 1980, and XRF analyses in Oct 1981.

TABLE III. INTERCOMPARISON OF SOURCE APPORTIONMENT RESULTS ($\mu g/m^3$) FOR DAYTIME, SEPT 10, 1980[a]

Size Range	Category	ERT-A CMB(EV)	ERT-B CMB(EV)	NEA CMB(EV)	TACB MLR	UI TTFA	SDSM CMB(XRD)	SUNY SAX	Microlab LM
Fine	Marine	0.58	0.53						
Fine	Crustal	3.9	1.9	1.7					
Fine	Vehicle Exhaust	3.8	2.3	3.7	11.5	17.6	0.46		
Fine	Steel	1.9	1.8	0.74					
Fine	Metals[b]			0.11					
Fine	Sulfate	23.4	25.6	25.8	41.1		50.8		
Fine	Carbon	6.8	8.8	7.7		36.3			
Fine	Nitrate								
Fine	Total	40.5	40.9	39.8	52.6	53.9	51.3		
Coarse	Marine	0.26	0.23	0.27		0.33	<0.01	0.0	
Coarse	Crustal	17.0	24.3	31.4	39.9	40.3	42.1	28.9	30.6
Coarse	Vehicle Exhaust	0.69	0.42	0.95			0.05	0.3	
Coarse	Steel		0.12	0.33				3.1	3.0
Coarse	Metals[b]			0.04				0.9	
Coarse	Sulfate	1.4	1.0	1.1		2.5		0.6	4.7
Coarse	Carbon	1.1	1.6	3.1			1.1	0.3	
Coarse	Nitrate	1.3	0.57	1.0		0.57			
Coarse	Total	21.6	28.2	38.3	39.9	43.7	43.3	34.1	38.4

[a] The mass concentrations measured in the fine and coarse fractions were 56.3 ± 2.8 and 38.4 ± 2.8 $\mu g/m^3$, respectively.

[b] Nonferrous metals.

TABLE IV. SOURCE APPORTIONMENT OF CRUSTAL MATTER ($\mu g/m^3$) IN THE COARSE FRACTION FOR DAYTIME, SEPT. 10, 1980

Crustal Matter	LBL XRD	SUNY SAX	Microlab LM
Road dust	a	14.5	a
Construction	a	3.9	a
Clay	a	a	10.6
Quartz	4.9	a	4.4
Ankerite	0.75	a	a
Gypsum	0.91	a	0.19
Calcite	4.7	2.0	3.5
Industrial calcium oxide and carbonate	a	a	2.1
Cement	a	0.3	0.46
Lime kiln	a	2.4	a
Slag	a	3.3	2.5
Coal fly ash	a	2.2	a
Oil fly ash	a	0.3	a
Fused fly ash	a	a	2.1
Glass	a	a	1.4
Minerals, misc.	0.19	a	0.38
Total crustal	11.5	28.9	27.6

[a] Not reported.